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# Microfibrillated Cellulose from Banana Stem Waste for High-Performance Bio-Hydrogels

***Linh Dinh Mai[1], Hung Tran Gia[1], Vinh Le Thi[2], Anh Nguyen Thi Quynh[2], Tung Nguyen Huy[2], Hieu Dao Minh[2], Hoanh Ngo Van[3], Mai Phan Thi Tuyet[2,*]***

[1]*School of Chemistry and Life Sciences, Hanoi University of Science and Technology, Ha Noi, Vietnam*
[2]*VNU University of Science, Vietnam National University, Ha Noi, Vietnam*
[3]*Institute of Chemistry and Materials, Ha Noi, Vietnam*
[*]*Corresponding author email: phanthituyetmai@hus.edu.vn*

**Abstract**

*In this work, microfibrillated cellulose (MFC) was successfully extracted from banana stem waste via an integrated chemical process of alkaline pretreatment with 0.5 M NaOH solution at 90 °C for x h (x = 2, 4, 6, 8 h), bleaching with 5% wt. $H_2O_2$ solution at 80 °C for 2 h, and acid hydrolysis with 4 M $HNO_3$ solution at 70 °C for 8 h, respectively. Obtained MFC samples were characterized by Scanning Electron Microscopy (SEM), Fourier Transform Infrared (FTIR), and X-ray Diffraction (XRD). The results confirmed that increasing alkali-treatment time from 2 h to 8 h raised the crystalline index from 66.1% to 84.3%, respectively, and revealed morphological transformation from hollow to ribbon fibers. Notably, the stable MFC suspensions were obtained at 2 h and 8 h, highlighting the potential of banana stem waste as a material for manufacturing MFC. These MFC samples were used as a performance enhancer for bio-hydrogel films. Adding MFC significantly increased the mechanical properties and water vapour permeability (WVP) of the alginate-based hydrogel film. Results also revealed that the performance of the bio-hydrogel films was strongly dependent on the morphology and crystallinity of MFCs: MFC hollow fibers with low CrI favored vapor permeability, whereas MFC ribbon fibers with high CrI improved mechanical strength. The obtained results have demonstrated the potential conversion of banana stem waste to value-added products, MFC, and high-performance bio-hydrogel for food packaging applications.*

Keywords: Alkali treatment time, banana stem waste, bio-hydrogel, crystallinity index, hollow fiber, microfibrillated cellulose.

## 1. Introduction

Microfibrillated cellulose (MFC), a semi-crystalline cellulose derivative, has gained significant attention for its excellent mechanical and barrier properties, which make it suitable for applications in the food, cosmetics, and medical industries [1, 2]. The incorporation of MFC from a variety of biomass sources into biopolymer matrices has been shown to enhance tensile strength and elastic modulus, while reducing elongation at break and water vapor permeability [3, 4]. Therefore, developing novel MFCs that can enhance the performance of biopolymer matrices has gained more and more interest from researchers. Commercial MFC is currently costly since it is made primarily on an industrial scale from wood or cotton sources. In contrast, agricultural residues represent an available and renewable source of lignocellulosic biomass, offering a sustainable material for diverse bioproducts. In recent years, significant efforts have been devoted to extracting MFC from agricultural wastes such as corn, rice straw, sugarcane bagasse, coconut, banana stem, jute, and pineapple leaves [5-13]. Among these, banana stem waste stands out as a promising cellulose source [8-11]. Bananas are cultivated globally as a major export crop, and each hectare generates approximately 50 tons of stem waste annually, posing serious disposal and environmental challenges [10]. Due to its high cellulose content (by dry weight), along with a porous fibrous architecture and loosely organized lignocellulosic matrix, banana stem biomass facilitates the efficient isolation of MFC, rendering it a promising feedstock for value-added applications. Previous studies have reported the extraction of cellulose microfibrils from banana stems with high crystallinity using sulfuric acid hydrolysis [11]. While high-crystallinity cellulose is desirable in some applications requiring high mechanical strength, it limits reactivity and restricts its suitability for the production of biohydrogel, bioplastic, and other green materials [5-8]. Pretreatment is a crucial step in biomass conversion, as it improves cellulose accessibility and reactivity [10, 11]. Alkaline treatment, one of the most widely used mercerization methods, effectively removes impurities and induces fiber swelling, thereby altering structural, morphological, and mechanical properties [14]. Previous research has shown that alkaline treatment can reduce both crystallinity degree and crystallinity index by disrupting the structure of fibers [15-18]. Despite these insights, little attention has been paid to the effect of alkaline treatment time on the extraction of MFC from banana stem waste. To fill this

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gap, the current study examines how the duration of alkaline treatment affects the crystal structure and morphology of MFC derived from banana stem by employing X-ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) techniques. Specifically, the crystallinity index and morphology of MFC fibers were evaluated to clarify the structural evolution during treatment.

Using hydrogel film as a packaging material to preserve food is a modern technology that is becoming a new trend in the world [1]. Synthetic polymer-based hydrogel films have been studied and applied in recent times [2]. But due to the lack of desirable biological properties, the trend towards gradually using natural polymer-based hydrogel (bio-hydrogel) films is attracting a lot of attention [3]. Natural polymers have been used to fabricate bio-hydrogel films, including chitosan, alginate, starch, pectin, cellulose, and their derivatives. Bio-hydrogel films made from cellulose and alginate have recently garnered significant research and application attention because the film formation process using an aqueous solvent does not affect the quality of the preserved products or pollute the environment [19, 20]. Furthermore, cellulose and alginate are the most abundant natural polymers. Although alginate-based bio-hydrogel films exhibit excellent biocompatibility and biodegradability, which make them attractive candidates for food packaging applications, their mechanical strength and water vapor permeability remain insufficient for practical use. Enhancing these functional properties through the incorporation of MFC with tailored morphology and crystallinity has therefore emerged as a key research focus. In the present study, the morphological characteristics and crystallinity of MFC fibers isolated from banana stems were modulated through controlled alkalization treatment to enhance the mechanical performance and water vapor permeability of alginate-based bio-hydrogel films. By valorizing banana stem waste as a cost-effective cellulose source, this study contributes to sustainable waste management while advancing the development of high-performance bio-hydrogels for food packaging applications.

## 2. Methodology

### *2.1. Materials*

The fresh banana stem waste (BSW) was collected from the Banana Farm, Ha Noi, Vietnam. The fresh banana stem was washed with water to remove dirt, then chopped into small pieces of about 0.5–1.0 cm using a grinding machine and subsequently dried in a hot air oven at 60 ºC for 24 h. The dried BSW powder was stored in zipper polyethylene bags.

This study uses nitric acid 65%, sodium hydroxide 99.5%, citric acid 99%, calcium chloride 99%, sorbitol 99% (Merck), and sodium alginate 98% (Himedia, India), sodium carboxymethyl cellulose isolated from pineapple leaves was obtained according to our previous study (average molecular weight (Mw) of 52,500 and degree of substitution (DS) of 0.9) [17], hydrogen peroxide 50% (Vietnam), as well as ethanol 99.9% from Xilong Chemical. They were of high purity.

### *2.2. Methods*

#### *2.2.1. Microfibrillated cellulose extraction*

The process of MFC extraction from BSW is depicted in Fig. 1. The dried BSW powder was first subjected to alkaline treatment with a fiber-to-liquid ratio of 1 g/25 mL using 0.5 M NaOH solution at 90 °C for 2–8 h while stirring vigorously. The solid residue was separated by vacuum filtration and thoroughly washed with warm RO water until pH 7. The resulting solid was bleached with 5 wt% $H_2O_2$ at a fiber-to-solution ratio of 1 g /20 mL at 80 °C for 2 h under continuous stirring. The bleached fibers were separated by vacuum filtration, then repeatedly washed with RO water to neutrality. Subsequently, acid hydrolysis was performed using 4 M $HNO_3$ at a fiber-to-liquid ratio of 1 g/25 mL at 70 °C for 8 h with vigorous stirring. The suspension was allowed to settle, the supernatant was decanted, and RO water was added; this washing–decantation cycle was repeated until pH 7. Finally, the neutralized suspension was centrifuged at 5000 rpm for 1 h to obtain MFC in paste form. The product was stored in sealed glass containers before characterization, and dried MFC powder was obtained by freeze-drying for further analyses. The stability of the MFC suspension was evaluated by allowing the dispersion to stand quiescently for 1 h, followed by visual observation of any sedimentation of solid particles at the bottom of the container.

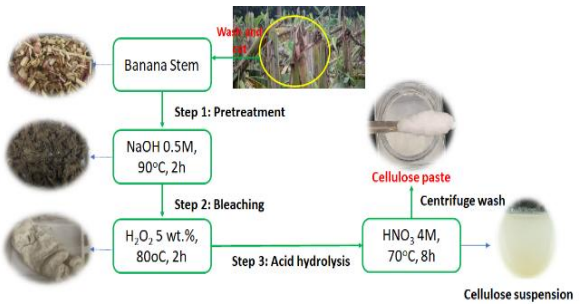



Fig. 1. Schematic illustration of the MFC extraction process from banana stem waste

The MFC extraction yield was determined according to (1):

$$H(\%) = \frac{m}{m_0} \times 100 \quad (1)$$

where $m_0$ is the weight of the dried banana stem powder, and $m$ is the weight of the dried MFC.

The concentration of MFC paste is also determined according to equation (1), in which $m_0$ and $m$ are the weights of MFC paste and dried MFC, respectively.

*2.2.2. Preparation of a hydrogel film*

The hydrogel films based on MFC, sodium carboxymethyl cellulose (CMC), and sodium alginate (alginate) were prepared using the solvent-casting method. Initially, MFC suspension (0.5 w/v) was prepared by adding MFC paste to 100 mL of distilled water and stirring vigorously at room temperature for 20 minutes. Then, alginate solution (2% w/v) was prepared by adding alginate powder (2.0 g) to 100 mL of distilled water and then stirring thoroughly until dissolution. At the same time, the CMC solution (2% w/v) was also prepared by adding CMC powder (2.0 g) to 100 mL of distilled water and then stirring thoroughly until dissolution. MFC suspension was then poured into the mixture of alginate solution and CMC solution at a ratio of dried weight of MFC/CMC/alginate of 20/60/20 (corresponding to a ratio of MFC/CMC/alginate solution volume of 4/3/1), and mixed thoroughly at 70 °C for 60 minutes to obtain MFC/CMC/alginate suspension. After adding citric acid (1% by mass) and $CaCl_2$ (1% by mass), the reaction was performed at 70 ºC for 90 minutes, stirring thoroughly at a speed of 1000 rpm. Finally, this mixture suspension was cast into a 9 cm diameter Petri dish, then dried at 60 °C for 8 h in the hot air oven. Hydrogel films were prepared based on two types of MFC, MFC2h and MFC8h, labeled as B-MFC2h and B-MFC8h, respectively. A hydrogel film based on alginate and CMC was also produced as a reference, labeled as Alginate. The network formation of the alginate and CMC hydrogel incorporated with MFC is described, as shown in Fig. 2.

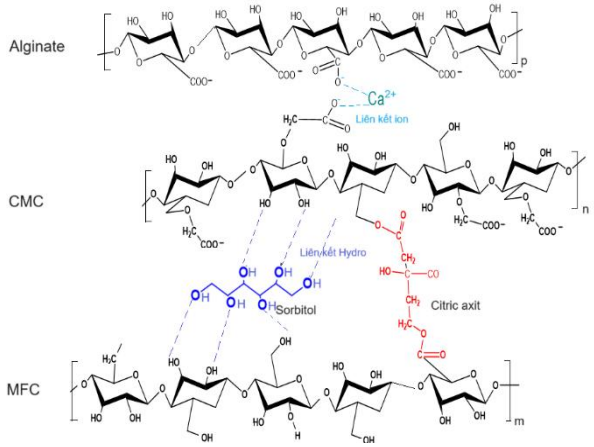



Fig. 2. Diagrammatic representation of the MFC-CMC-alginate hydrogel film's network creation

*2.2.3. Fourier Transform Infrared Spectroscopy*

FTIR analysis of the MFC samples was conducted using a FT/IR-6300 spectrometer with the KBr pellet technique. The spectral resolution was 4 $cm^{-1}$, and the absorption range was 600–4000 $cm^{-1}$.

*2.2.4. X-ray diffraction*

X-ray diffraction analyses were conducted in ambient conditions, utilizing a Shimadzu XRD-6100 diffractometer with CuKα radiation ($\lambda$=1.54Å). The angle of diffraction varied between 5 and 80° (0.05°/min). The measurement was carried out at 30 kV and 15 mA using Cu $K_\alpha$ radiation. The crystallinity index (CrI) of the obtained MFC samples was evaluated by Segal methods using (2), respectively.

The crystallinity index of the samples was calculated by following the equation:

$$CrI\ (\%) = (I - \frac{I_{am}}{I_{002}}) \times 100 \qquad (2)$$

where $I_{002}$ is the maximum intensity of the peak corresponding to the plane in the sample with the Miller indices (002) at a $2\theta$ angle between 22–23°, indicating crystalline material, and $I_{\mathrm{am}}$ is the intensity of diffraction of the non-crystalline material, which is taken at an angle of approximately $2\theta$ angle of 18° in the valley between the peaks; the minimal intensity represents amorphous material in cellulose fiber [21, 22].

*2.2.5. Scanning electron microscopy*

The SEM images show the surface of the MFC samples. The SEM images were acquired using a Hitachi Tabletop Microscope TM4000Plus operated in BSE mode at an accelerating voltage of 15 kV. The open-source software ImageJ was used to measure image dimensions in SEM. All MFC samples were dried using an Alwhales Freezing Dryer at -75 ºC and 20 Pa.

*2.2.6. The thickness and mechanical properties*

A digital micrometer (Mitutoyo, Japan) was used to measure the thickness of the hydrogel layer with a 1µm accuracy. Using a LLOYD machine that was operated with an initial distance between grips of 50 mm and a crosshead speed of 50 mm/min, the mechanical characteristics of hydrogel films were assessed in accordance with the ASTM D882-18 standard procedure [23]. Measurements were made of the tensile modulus, elongation at break, and tensile strength. Before testing, the hydrogel films were cut into rectangular strips of 10 x 100 mm and conditioned for 48 hours at 25 °C and 50% humidity. Five replications of each test were conducted, and the findings are shown as averages of these data.

*2.2.7. Water vapour permeability*

The permeability of hydrogel films to water vapor was measured gravimetrically using a Water vapour permeability (WVP) cup following the ASTM 96 standard method [2]. Before testing, hydrogel film samples were stored at 50% RH and 25 ºC. The WVP cup was filled with silica gel beads, then covered with the hydrogel film, sealed, and kept in a controlled environmental desiccant (25 ºC and 75% RH). The experiment was carried out over 7 days at RH of

75 ± 3%. The WVP cup's weight was measured at regular time intervals, and the weight addition was determined. The WVTR ($g/m^2.s$) was determined from the slope of the steady-state portion of the weight addition of the cup versus the time curve. The WVP ($g.mm/kPa.day.m^2$) of the hydrogel film was calculated by the following equation:

$$WVP = (WVTP*L)/p \quad (3)$$

where $L$ was the film thickness (mm), and $p$ was the water vapor partial pressure difference (kPa) across the film.

*2.2.8. Water vapor absorption*

Test samples in the form of 9 $cm^2$ pieces were dried to constant weight according to the following procedure: the samples were dried at 105 ºC for 1 h, then cooled in a desiccator for 15 min and weighed. This drying procedure was repeated until the difference between the two weightings was not more than 0.01%. Samples were stored in a desiccator.

The test samples were placed in different stable humidity environments: 50±3%, 75±3%, and 97±3%, named RH1, RH2, and RH3, respectively. Samples were weighed after 48 h. The water vapor absorption capacity of the film was calculated by the following equation:

$$Q = \frac{m_2 - m_1}{m_1} x100, \% \quad (4)$$

where: $m_1$(g) and $m_2$ (g) are the weights of the dry and moistened samples, respectively.

Data processing and graphical analysis were carried out using Microsoft Excel and Origin software.

## 3. Results and Discussion

### *3.1. Extraction of Microfibrillated Cellulose from Banana Stem Waste*

The effect of alkaline treatment time on the extraction of MFC from banana stem waste was investigated.

Table 1 presents the state of suspensions, concentration of pastes, extracted yields, and crystallinity of extracted MFC samples. The state of MFC2h suspension and MFC8h suspension remained stable, whereas MFC4h suspension and MFC6h suspension showed sedimentation. With increasing alkali treatment time from 2 h to 6 h, the paste concentration increased, but it decreased sharply at 8 h. The concentration of paste reflects the size of MFC fibers, with lower values indicating smaller fiber dimensions. Notably, the MFC4h and MFC6h samples exhibited higher concentrations than the MFC2h sample.

Fig. 1 shows photographs of the obtained MFC in the paste and suspension.

The morphology of the obtained MFC is presented in Fig. 3. SEM images reveal clear changes in the shape and size of MFC fibers according to the increase in alkali treatment time. The MFC2h sample exhibited hollow fibers with an average diameter of 20 μm. After an alkali treatment time of 4 h, the fibers retained their hollow structure but became smoother and smaller, with diameters in the range of 8–15 μm. When increasing the alkali-treatment time to 6 h, the hollow fibers were broken into ribbon-like structures with a width of ~25 μm. When increasing the alkali-treatment time to 8 h, the ribbon fibers became smaller, with a width of 2–10 μm. These results demonstrated a morphological transition from hollow to ribbon fibers as the alkali-treatment time increased from 4 h to 6 h, resulting in a decrease in the water retention capacity of the fibers, leading to an increase in the paste concentration. This finding is consistent with the increase in MFC paste concentration at the alkali-treated times of 4 h and 6 h compared to 2 h in Table 1.

In this study, the crystalline characteristics of the obtained MFC samples were analyzed using XRD [19, 20]. The XRD patterns of the MFC samples are presented in Fig. 4. The XRD patterns of all MFC samples derived from banana stems exhibited three characteristic peaks at $2\theta$ equal to 15.6°, 22.4°, and 34.6°, corresponding to the (110), (002), and (004) planes of cellulose crystals [8, 10, 19, 20]. The crystallinity indices calculated from the XRD data (Table 1) increased with longer alkali treatment times, indicating progressive removal of lignin and hemicellulose.

Table 1. Parameters of cellulose extraction at different alkali treatment times

| **Parameters** | **MFC2h** | **MFC4h** | **MFC6h** | **MFC8h** |
|---|---|---|---|---|
| State of MFC suspension solution | stable | settle | settle | stable |
| MFC paste concentration, % w/w | 3.4 | 4.3 | 6.5 | 2.5 |
| CrI, % | 66.1 | 78.5 | 81.7 | 84.3 |
| H, % | 48.6 | 48.1 | 47.2 | 47.8 |

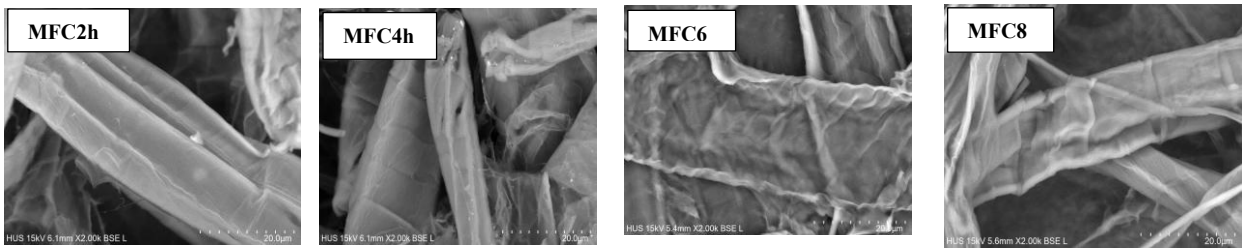



Fig. 3. SEM images of banana stem MFC samples at different alkali treatment times at 2000 x magnification (20 µm size bar)

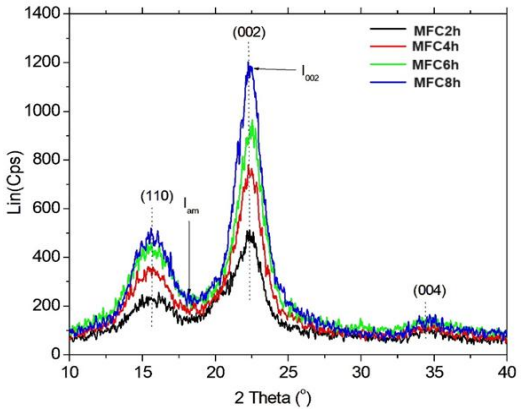



Fig. 4. XRD patterns of MFC samples at different alkali treatment times

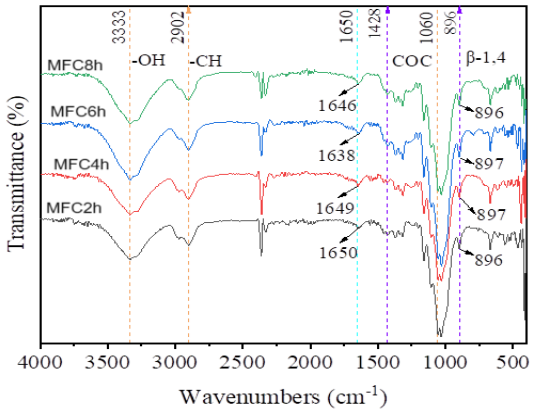



Fig. 5. FTIR spectra of MFC samples at different alkali treatment times

In this study, prolonging the alkaline treatment resulted in a progressive increase in the crystallinity index of the extracted MFC. This behavior contrasts with the findings of Mittal *et al.* [17], who reported a marked decrease in crystallinity due to the conversion of cellulose I to cellulose II under highly concentrated NaOH treatment (16.5 wt%, ~4.125 M). The observed discrepancy can be rationalized by considering the substantial difference in alkali concentration and its consequent impact on the cellulose supramolecular structure. Under the strong alkaline conditions employed by Mittal *et al.*, the native crystalline lattice of cellulose was sufficiently disrupted, inducing polymorphic transformation and increased structural disorder. In contrast, the comparatively mild alkaline conditions applied in the present work (0.5 M NaOH) are not severe enough to trigger allomorphic conversion. Instead, they predominantly promote the selective removal of amorphous constituents, including lignin and hemicellulose. Consequently, the relative proportion of crystalline cellulose in the solid fraction increases, leading to an apparent increase in the crystallinity index of the MFC samples.

The crystallinity index (CrI) of the MFC2h sample is 66.1, notably lower than 82.7 reported by Maha *et al.* [14]. As we know, the crystallinity of cellulose is influenced by the method used for extraction and treatment. Therefore, the extraction technique applied in this study gives cellulose with comparatively low crystallinity.

The FTIR spectra of the obtained celluloses are presented in Fig. 5. In the MFC spectra, a wide band at 3333 $cm^{-1}$ is linked to O–H stretching, whereas the peak at 2902 $cm^{-1}$ is assigned to C–H stretching vibrations. The peaks at 1367 and 1428 $cm^{-1}$ are ascribed to the bending vibrations of C–H and C–O in the rings of polysaccharides, respectively. The C–O group vibration in the cellulose backbone's secondary alcohols is observed at 1129 $cm^{-1}$. Bands in the range of 896–1051 $cm^{-1}$ are associated with C–O groups linked to β-(1→4)-glycosidic bonds between glucose units [14-18, 24]. These features are consistent with previous reports by Kumari *et al.* [12]. Notably, the band at 896 $cm^{-1}$, corresponding to the amorphous region of cellulose, decreased progressively with increasing alkali treatment time, in agreement with the XRD results. The absence of the peak at 1735 $cm^{-1}$, attributed to carbonyl groups of aromatic acids in lignin and hemicellulose, confirms the removal of these components and the purity of the obtained MFC.

Table 1 summarizes the cellulose extracted yield at different alkali treatment times with 0.5 M NaOH. The yield remained nearly constant with treatment time, reaching 48.6% at 2 h, which is higher than values reported for rice straw (32 wt.%) [16], but slightly lower than that from pineapple leaf (51 wt.%) [17]. Prolonged alkali treatment (2–8 h) did not markedly affect yield but significantly altered fiber morphology, shifting from hollow to ribbon structures. The MFC suspensions obtained at 2 h and 8 h were stable and suitable for

bio-hydrogel film preparation. Crystallinity also increased with treatment time, from 66.1% (2 h) to 84.3% (8 h).

As mentioned above, MFC has gained increasing importance as a sustainable material for the fabrication of high-performance and biodegradable materials (bio-hydrogels, bioplastics, and related biomaterials). Cellulose crystallinity and structural order, strongly influenced by both its botanical origin and extraction method, play a critical role in determining its chemical reactivity. Substitution reactions predominantly occur in the amorphous domains of the cellulose structure. Consistent with this understanding, Mhenni *et al.* [15] reported that an increase in cellulose crystallinity leads to a reduced reactivity toward carboxymethylation. For enhanced chemical reactivity of MFC, a lower degree of crystallinity is advantageous. Moreover, MFC in paste form is preferred over the dried state, as drying often induces irreversible fiber aggregation, which significantly hinders redispersion before use. Therefore, considering the combined requirements of low crystallinity and suspension stability, an alkaline treatment time of 2 h was identified as the most suitable condition for producing MFC in this study. Therefore, isolated MFC from banana stems was used as a material to enhance alginate-based hydrogel film, and its effects on the tensile properties and water vapor permeability of hydrogels containing hollow and ribbon fibers were evaluated.

### *3.2. Characterization of Hydrogel Films*

In this work, hydrogel films were analyzed under 4 headings as water vapor permeability, mechanical strength, moisture absorption, morphology, and structural features.

The water vapor permeability is the water vapor transmission with diffusion of a material, and it is important for the preservation of food, especially fresh fruits. The results of the vapor permeability of the bio-hydrogel film samples are presented in Table 2.

Table 2. WVP value of bio-hydrogel film samples

| **Sample** | B-MFC2h | B-MFC8h | Alginate |
|---|---|---|---|
| **WVP, g.mm/kPa.day.m$^2$** | 18.9±0.6 | 13.5±0.4 | 4.9±0.3 |

The results showed that the WVP of bio-hydrogel films containing MFC in both types, B-MFC2h and B-MFC8h, was significantly higher than that of the control alginate sample, with values of 18.9, 13.5, and 4.9 g.mm/kPa.day.m², respectively. The greater WVP of hydrogel films with the presence of MFC indicated that MFC may have contributed to the formation of a more rigid network structure for the hydrogel film, thereby increasing porosity and facilitating the easier permeation of water vapor molecules. In addition, this result also reflected that MFC bonds to water vapor molecules more difficultly than CMC and alginate, as reported by Xiao Zhang *et al.* [25]. This can be explained by the fact that MFC has a higher crystallinity than alginate and CMC, resulting in fewer free -OH groups on the fiber surface, which reduces the interaction between the MFC fiber surface and water molecules, ultimately allowing water vapor molecules to permeate through the film more easily. Notably, the hydrogel film with MFC in hollow fibers showed significantly higher WVP than that with MFC in ribbon fibers. This can be explained by the fact that the MFC in hollow fibers allows vapor to pass through the film more easily, resulting in increased vapor permeability. The markedly enhanced water vapor permeability of the MFC-based bio-hydrogel films underscores their potential as advanced packaging materials for food preservation, particularly for fresh fruits and vegetables that remain metabolically active and continue to respire postharvest.

The mechanical strength of the bio-hydrogel film samples is presented in Table 3.

Table 3. Mechanical strength of bio-hydrogel films

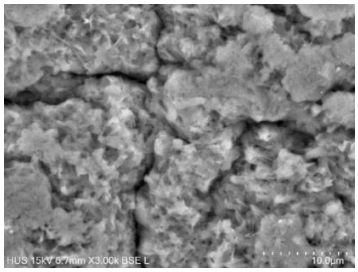
| **Sample** | **σ, MPa** | **E, MPa** | **ε, %** | **δ, µm** |
|---|---|---|---|---|
| Alginate | 8.5±0.4 | 5.4±0.3 | 157±5 | 51±3 |
| B-MFC2h | 13.9±0.4 | 11.2±0.3 | 120±5 | 52±2 |
| B-MFC8h | 18.5±0.4 | 14.3±0.3 | 125±5 | 47±2 |

It can be observed that the bio-hydrogel films containing MFC extracted from banana stem, in both types (B-MFC2h and B-MFC8h), exhibited significantly higher tensile strength ($\sigma$) and tensile modulus ($E$) compared to the control. In contrast, the elongation at break ($\varepsilon$) of the MFC-containing hydrogel films decreased relative to the control. The thickness ($\delta$) of the hydrogel film samples was in the range of 47 to 52 µm. This result can be explained by the presence of MFC molecular chains within the hydrogel film structure, which enhances the mechanical strength of the three-dimensional network, restricts chain mobility, and consequently leads to increased tensile strength and reduced elongation. Furthermore, the bio-hydrogel films containing MFC ribbon fibers exhibited a significant increase in mechanical strength compared to those with hollow fibers.

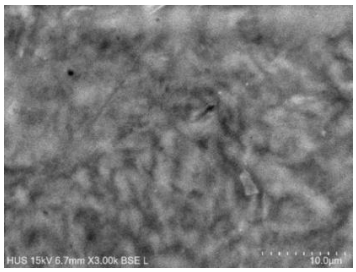
The results of the moisture absorption of the bio-hydrogel film samples are shown in Table 4.

Table 4. The moisture absorption value of the bio-hydrogel films in different humidity environments

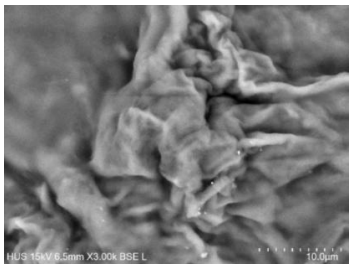
| Humidity environment | Q, % | | |
|---|---|---|---|
| | Alginate | B-MFC2h | B-MFC8h |
| RH1 | 31±3 | 53±3 | 44±3 |
| RH2 | 57±8 | 112±5 | 87±5 |
| RH3 | 95±10 | 205±13 | 155±11 |



(a)



(b)



(c)

Fig. 6. SEM images of bio-hydrogel films: (a) Alginate, (b) B-MFC8h, and (c) B-MFC2h, at 3000x magnification (10nμm size scale)

In contrast to the alginate control sample, the B-MFC2h and B-MFC8h bio-hydrogel films exhibit noticeably higher moisture absorption. After 48 hours of absorption in the RH1 humidity, the moisture absorption reached 53±3%, 44±3%, and 31±3% for B-MFC2h, B-MFC8h, and alginate samples, respectively. It can be seen that the moisture absorption capacity of the bio-hydrogel film samples increased sharply when the humidity increased. The results in Table 1 show that the moisture absorption of the B-MFC2h bio-hydrogel film sample reached 53±3%, 112±7%, and 205±13% for RH1, RH2, and RH3, respectively. Remarkably, as the humidity environment increased, the increase in the water vapor absorption of the bio-hydrogel film sample incorporating MFC hollow fibers (B-MFC2h) was significantly higher than MFC ribbon fiber-based bio-hydrogel film sample (B-MFC8h). Therefore, this result reflected that MFC's morphology and structural features also significantly affect the moisture absorption capacity of the bio-hydrogel film. The marked improvement in the hydrogel film's moisture absorption by adding MFC extracted from banana stem waste could be explained by the formation of a strong network structure based on MFC frameworks. These valuable results confirmed that the incorporation of MFC extracted from banana stems in the alginate-CMC matrix enhanced the moisture absorption capacity of the bio-hydrogel film

To examine the morphology of hydrogel films, SEM images are presented in Fig. 6.

As shown in Fig. 6a, alginate films have a semi-rough surface with noticeable patches of high roughness and fractures, whereas a smooth surface is observed across the entire surface of the B-MFC2h bio-hydrogel film (Fig. 6b). In the case of the B-MFC2h bio-hydrogel film (Fig. 6c), the surface roughness shows numerous pores but no surface fractures. The bio-hydrogel film's surface has become smoother and free of damaged areas due to the presence of MFC. The irregular surface with some fractured patches observed in the alginate film can be explained by weakening or even loss of bonding, resulting in a less intact film structure. Consequently, incorporating MFC into the alginate-CMC network altered the morphological characteristics and surface integrity of the bio-hydrogel film. Prior research has never documented these distinctive surface features. As a result, cellulose hollow fibers were used to create an MFC-alginate bio-hydrogel with higher porosity because of the internal network. Furthermore, the number of pores on the surface of B-MFC2h increased when adding MFC hollow fibers, which in turn increased the direct pathways for water vapor to penetrate through the film. Thus, SEM image results demonstrated that the improvement in surface morphology of the MFC bio-hydrogel films led to an increase in mechanical strength and water vapor permeability for both types of MFC, as hollow fibers and ribbon fibers. Notably, the change in morphology of the hydrogel film was completely consistent with the change in thickness of the bio-hydrogel film given in Table 3. The addition of MFC into the alginate-CMC network increased the thickness of the bio-hydrogel film for MFC hollow fibers while decreasing the film thickness for MFC ribbon fibers.

The XRD patterns of the B-MFC2h, alginate bio-hydrogel, and MFC2h (labeled MFC) samples are presented in Fig. 7.

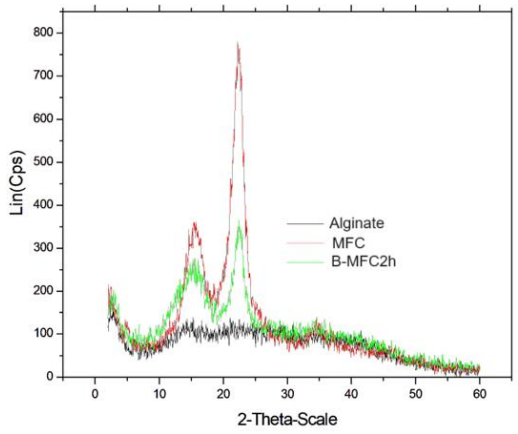



Fig. 7. XRD patterns of bio-hydrogel film and MFC samples

The alginate film exhibited no characteristic peaks within the 10–40° range, whereas the B-MFC2h sample showed distinct peaks at $2\theta$ equal to 15.6° and 22.4°, linking to the (110) and (002) planes of cellulose crystals. Its crystallinity index was calculated to be 15.6%. The appearance of characteristic peaks in the crystalline structure of the B-MFC2h microfibrillated cellulose-containing hydrogel film is due to the presence of MFC2h microfibrillated cellulose. Thus, MFC derived from banana stems altered the morphology of the alginate matrix, leading to increased tensile strength and water vapor permeability. Accordingly, the bio-hydrogel based on MFC isolated from banana stems exhibits improved mechanical performance and enhanced water vapor permeability, rendering it suitable not only for food packaging applications but also for broader uses, such as wound dressings and breathable films.

## 4. Conclusion

MFC was successfully extracted from Vietnamese banana stem waste with a maximum yield of 48.6 wt.% under combined alkaline treatment, bleaching, and hydrolysis treatments. The morphology and crystalline index (CrI) of MFC were effectively tuned by alkali treatment time, shifting from hollow fibers with lower CrI (lowest of 66.1%) to ribbon fibers with higher CrI (up to 84.3%). A stable state of MFC suspension solution was also achieved with alkali treatment times of 2 h and 8 h. These results highlight banana stem waste as a promising and sustainable raw material for industrial MFC production.

MFC was extracted from Vietnamese banana stem waste, with morphology and structure influenced by alkalinization time: 2 hours produced hollow fibers with low crystallinity, while 8 hours resulted in ribbon fibers with high crystallinity.

MFC product enhanced the mechanical strength, water vapor permeability, and absorption of Bio-Hydrogel films. Bio-Hydrogel film performance was influenced by MFC morphology and crystallinity; hollow fibers with low crystallinity favored vapor permeability and moisture absorption, while ribbon fibers with high crystallinity improved mechanical strength.

MFC extracted from banana stem waste significantly improved the mechanical strength, water vapor permeability, and moisture absorption of alginate-CMC-based biohydrogel films. The performance of the film was strongly dependent on the morphology and crystallinity of MFC: MFC hollow fibers with low CrI favored vapor permeability and moisture absorption, whereas MFC ribbon fibers with high CrI improved mechanical strength. These findings highlight banana stem waste-derived MFC as a promising material for the development of high-performance bio-hydrogels.

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